

THE STRENGTHENING OF ENERGETIC MATERIALS UNDER PRESSURE

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ABSTRACT

The mechanical properties of composite plastic bonded explosives are being studied as a function of hydrostatic confining pressure. The flow stress and the modulus obtained in simple compression are both found to increase with increasing confining pressure. Thus, these materials become stronger and stiffer with increasing pressure. The sensitivity to pressure, however, decreases with increasing pressure. The failure process also changes with pressure. At atmospheric pressure failure is primarily due to crack processes while at elevated pressures failure is due primarily to plastic flow. Several mechanisms which may account for the pressure dependencies of the flow stress, the modulus and the failure processes are discussed.

1. INTRODUCTION

Energetic materials are often used under conditions of mechanical confinement, e.g., explosives by the steel casings and propellants by the breach and the high pressures during burning. When modeling the response of energetic materials to planned and unplanned mechanical stimuli, it is necessary to know the mechanical failure modes and other mechanical properties as a function of confinement. Previously reported studies indicate a change with confinement in failure modes but not elastic properties for compression of polycrystalline explosives, i.e., TNT (trinitrotoluene) and Composition B, a composite of TNT and RDX (cyclotrimethylene trinitramine) (Wiegand et al., 1991). In addition, the yield strength observed with confinement is independent of confining pressure (Pinto and Wiegand, 1991). While studies of composite plastic bonded explosives also indicate a change in compressive failure mode with confinement, use of the same steel cylinder technique as used for TNT and Composition B indicates that the results cannot be interpreted in terms of properties independent of confining pressure (Mezgar, M. et al., unpublished results). The work reported here was undertaken to investigate the confining pressure dependence of failure and other mechanical properties of plastic bonded explosives.

2. EXPERIMENTAL

A high pressure chamber designed to contain pressures up to 138 MPa was used to study the compressive mechanical properties as a function of confining pressure (Wiegand, 2000). Hydraulic oil was used as the confining

medium and the sample in the form of a right circular cylinder was protected from the oil by a tight fitting tubular gum rubber or neoprene shroud. A sketch of the sample, shroud and sensors is given in Figure 1.

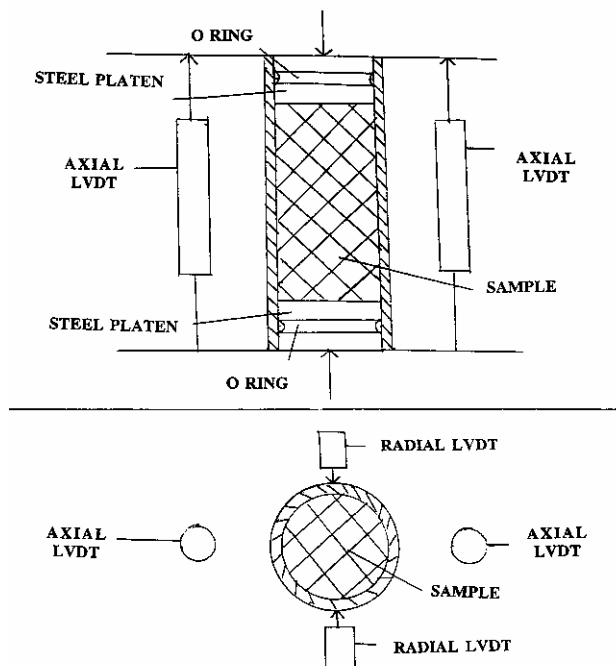


Figure 1 Side and end sketches of the sample, shroud and sensors for compression at constant pressure.

The ends of the sample were against steel platens and O-ring seals were used to prevent oil from reaching the sample. The samples were compressed along the cylindrical axis and two LVDT's (linear voltage differential transformers) were mounted to measure axial strains. They were spaced 180 degrees apart around the circumference of the sample with their axes parallel to the sample axis. The sample axial strain was taken as the average of the strains obtained from the two LVDT's. Two or in some cases three additional LVDT's were mounted to measure radial strains. The confining pressure is taken here as the cell hydrostatic pressure before the start of and/or during the axial compression. Measurements at atmospheric pressure were made in air.

Axial stress versus axial strain data in compression were obtained using the above chamber and an MTS servo-hydraulic system operated at a constant displacement rate (Wiegand et al., 1991; Pinto and Wiegand, 1991). Most of the work was carried out at a strain rate of approximately 0.001/sec. but some work was

Report Documentation Page			Form Approved OMB No. 0704-0188		
Public reporting burden for the collection of information is estimated to average 1 hour per response, including the time for reviewing instructions, searching existing data sources, gathering and maintaining the data needed, and completing and reviewing the collection of information. Send comments regarding this burden estimate or any other aspect of this collection of information, including suggestions for reducing this burden, to Washington Headquarters Services, Directorate for Information Operations and Reports, 1215 Jefferson Davis Highway, Suite 1204, Arlington VA 22202-4302. Respondents should be aware that notwithstanding any other provision of law, no person shall be subject to a penalty for failing to comply with a collection of information if it does not display a currently valid OMB control number.					
1. REPORT DATE 00 DEC 2004		2. REPORT TYPE N/A		3. DATES COVERED -	
4. TITLE AND SUBTITLE The Strengthening Of Energetic Materials Under Pressure			5a. CONTRACT NUMBER		
			5b. GRANT NUMBER		
			5c. PROGRAM ELEMENT NUMBER		
6. AUTHOR(S)			5d. PROJECT NUMBER		
			5e. TASK NUMBER		
			5f. WORK UNIT NUMBER		
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) U.S. Army RDECOM, ARDEC Building 3022, Picatinny, NJ 07806-5000			8. PERFORMING ORGANIZATION REPORT NUMBER		
9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)			10. SPONSOR/MONITOR'S ACRONYM(S)		
			11. SPONSOR/MONITOR'S REPORT NUMBER(S)		
12. DISTRIBUTION/AVAILABILITY STATEMENT Approved for public release, distribution unlimited					
13. SUPPLEMENTARY NOTES See also ADM001736, Proceedings for the Army Science Conference (24th) Held on 29 November - 2 December 2005 in Orlando, Florida.					
14. ABSTRACT					
15. SUBJECT TERMS					
16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT UU	18. NUMBER OF PAGES 7	19a. NAME OF RESPONSIBLE PERSON
a. REPORT unclassified	b. ABSTRACT unclassified	c. THIS PAGE unclassified			

done at strain rates up to 0.1/sec. The right circular cylinder samples were 3.81 cm (1.50 inch) in length and 1.90 cm (0.75 inch) in diameter and so had a length to diameter ratio of two. The end faces of all samples were coated with a lubricant to minimize frictional effects between the sample end faces and the loading platens. The sample temperatures during measurements were between 20 and 23 C and samples were conditioned at temperature for at least two hours before measurement. The dimensions of all samples at 0.1 MPa (atmospheric pressure) were used to obtain engineering stress and engineering strain.

Most of the results presented here of measurements in the high pressure chamber are for a composite, PBS 9501, containing 94% sucrose, an inert, and a binder (see Table 1).

Table 1

Composition of Explosive Composites
Binder

Name	Explosive/Inert	Polymer	Plastizer	T _g (°C)
PAX2A	HMX	CAB	BDNPA/F	-37 ^(a)
	85%	6%	9%	
PBS 9501	SUCROSE	ESTANE	BDNPA/F	-41(B) ^(b)
	94%	3%	3%	
PBX 9501	HMX	ESTANE	BDNPA/F	-41(B) ^(b)
	95%	2.5%	2.5%	
LX-14	HMX	ESTANE		-31(B) ^(c)
	95.5%	4.5%		
Composition B	RDX	TNT	WAX	
	59.5%	39.5%	1%	

Nomenclature: HMX - Cyclotetramethylene tetranitramine. RDX - Cyclotrimethylene trinitramine. NC - Nitrocellulose. CAB - Cellulose Acetate Butyrate. BDNPA/F - Bis(2,2-Dinitropropyl)Acetal/Formal. Estane - Polyurethane. B – Property of the Binder ((a) Harris, J., Private Communication; (b) Flowers, G. L., Private Communication; (c) Dobratz et al., 1985).

This composite was developed as an inert mechanical simulant for a plastic bonded explosive, PBX 9501, composed of 95% HMX and the same binder. The unconfined compressive mechanical properties of PBS 9501 are very similar to those of PBX 9501 (Funk et al., 1996). Some results of measurements in the high pressure chamber are also presented and/or discussed for PBX 9501 and two other composite plastic bonded explosives, LX-14 and PAX 2A (see Table 1). Samples of the composites were prepared by pressing or casting

(Composition B) into large billets and machining to size. Precautions were taken to insure that the cylinder end faces were adequately flat and parallel (Funk et al., 1996; Wiegand et al., 1991; Idar, D., private communication). The densities of all samples were in a narrow range close to the maximum theoretical (zero porosity) density.

3. RESULTS

In Figure 2 the compressive axial stress-strain response of PBS 9501 is given for several confining pressures (Wiegand, 2000 and Wiegand and Reddingius, 2004).

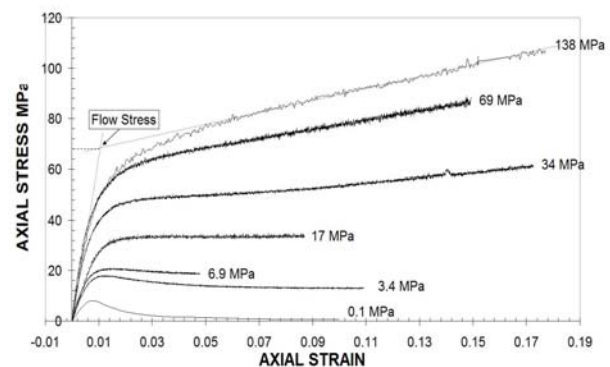


Figure 2 Axial stress versus axial strain for samples of PBS 9501 for confining pressures from bottom to top of 0.1 (atmospheric), 3.4, 6.9, 17, 34, 69 and 138 MPa.

There are significant differences between the curves for the lower confining pressures and the curves for the higher confining pressures. These include: a) a maximum stress for the lower confining pressures which is not observed at the higher confining pressures; and b) a change from strain softening after the maximum at the lower confining pressures to work hardening at larger strains at the higher confining pressures. In addition, the initial slope is larger at the higher confining pressures. Young's modulus is defined as the initial slope at atmospheric pressure. This initial slope is referred to here simply as the modulus at higher confining pressures.

A yield strength, taken at the point at which the initial part of the stress-strain curve deviates from linearity (Figure 2) has been found to be variable from sample to sample and is not considered here. To better characterize the data a flow stress is taken as the stress at the intersection of a straight line fitted to the work hardening part of the stress-strain curve with the straight line fitted to the initial modulus portion of the curve. This is indicated in Figure 2 for the data at 138 MPa and is the stress at which significant plastic flow occurs. For PBS 9501 this flow stress is numerically very close to the yield strength as obtained by a one percent strain offset method. The flow stress at the lower pressures is taken as the maximum stress.

As shown in Figure 3 measurements as a function of confining pressure indicate that the modulus increases at a continually decreasing rate as pressure is increased. Thus, the initial rate at the lowest pressures is more than an order of magnitude greater than the rate at the highest pressures.

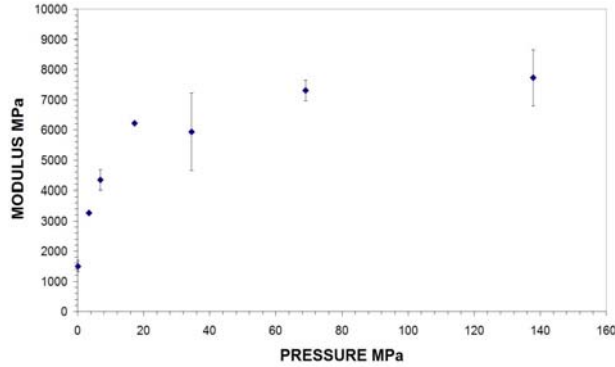


Figure 3 Modulus versus confining pressure for PBS 9501.

A similar rate of increase with pressure is found for the flow stress as shown in Figure 4.

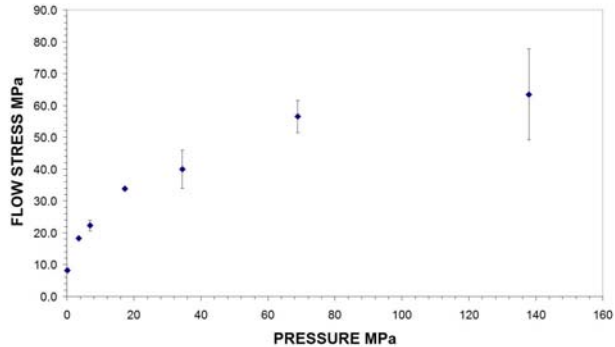


Figure 4 Flow stress versus confining pressure for PBS 9501.

However, there is too much scatter in the present data to determine if there is a simple relationship between the modulus and the flow stress as a function of pressure. The maximum slope of the stress-strain curve in the work softening/work hardening region increases from negative to positive values with increasing pressure and the rate of increase with increasing pressure also decreases. This is shown in Figure 5. Thus, the whole stress-strain curve becomes less sensitive to pressure at the higher pressures of Figure 2. There is considerable spread in the work hardening slope at 138 MPa as indicated by the large error bars of Figure 5 at this pressure. The error bars of Figures 3, 4 and 5 are the standard deviations of the measured values. In these figures each point is the average of the results for two or three samples with the exception of the points at 3.4 and 17 MPa which represent the results of only one sample. Increases in the yield strength and the modulus with increasing pressure have been reported for

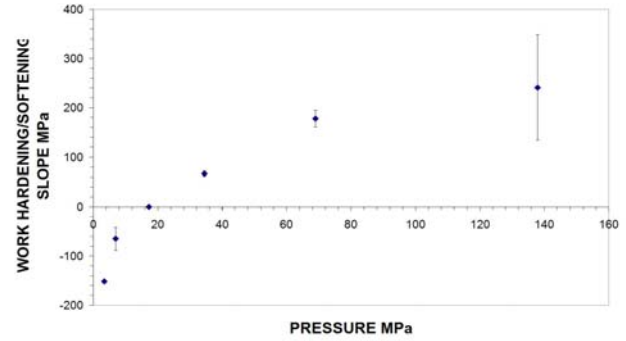


Figure 5 Work softening/work hardening slope versus confining pressure for PBS 9501.

polymers and polymer composites, including gun propellants (Hoppel et al., 1995; Constantino et al., 1985; Constantino et al., 1987; Ward and Hardley., 1993).

Results similar to those of Figures 2, 3, 4 and 5 were also obtained for LX-14 and very limited results for PBX 9501 suggests similar behavior as a function of pressure. Results similar to those of Figure 2 were also obtained for PAX 2A. While detailed results for these three composites will be published separately the stress-strain curves for unconfined and confined PAX 2A are given in Figure 6 (Wiegand, D. A., unpublished results).

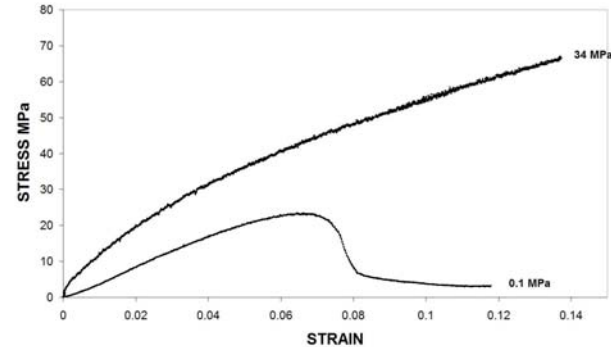


Figure 6 Axial stress versus axial strain for PAX 2A with confining pressures from bottom to top of 0.1 MPa (atmospheric) and 34 MPa.

For PAX 2A the stress-strain response has continuous curvature (see Figure 6) so that linear regions are not clearly identifiable. Therefore, the strain offset method is used to obtain a measure of the yield strength and modulus is taken as the initial slope of the stress-strain curves. The work hardening coefficient is taken as the average slope at larger strains.

The strain softening at 0.1 MPa (lowest curves of Figures 2, and 6 has been attributed to damage due to crack growth processes (Dienes, 1998; Wiegand, D. A., unpublished results). Therefore, the results of these figures suggest that this crack growth does not occur at the higher pressures where work softening is not

observed. Thus, there appears to be a shift from work softening due to crack growth at low confining pressures to work hardening and plastic flow at higher pressures. The photograph of Figure 7 shows pictorial evidence to support this postulate of a change in failure processes with increasing pressure.

Samples

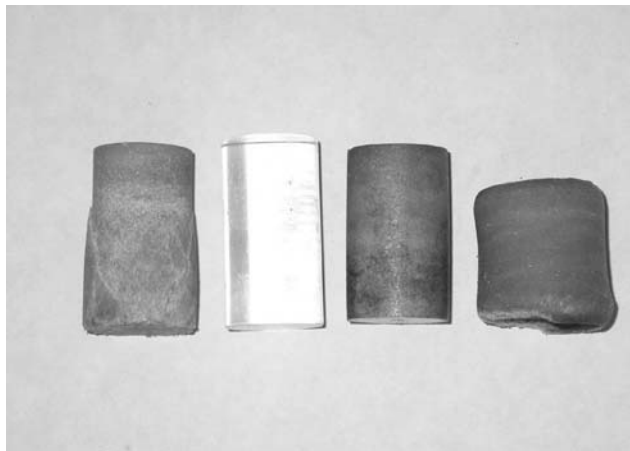


Figure 7 Photograph of deformed samples of PBS 9501 and a reference plastic sample of the same dimensions as the PBS 9501 samples before deformation. From left to right: sample compressed axially with a confining pressure of 0.1 MPa; plastic reference sample; samples compressed axially with confining pressures of 138 MPa, and 69 MPa. The maximum axial strain differs for each sample. The sample deformed at 138 MPa was graphite coated before deformation.

The sample compressed at 0.1 MPa shows extensive surface cracking while the sample compressed at 69 and 138 MPa show no evidence of surface cracking. The total axial strain was different for each sample of Figure 7 and it is clear from the figure that the retained or permanent axial strains also differ for each sample. The sample compressed at 0.1 MPa has, in addition to extensive cracking, a large radial expansion at the bottom but negligible radial expansion at the top. A gradient of radial strain is often observed for this type of sample, this amount of axial compression and this confining pressure (atmospheric). The permanent axial strain for this sample is -5.2%. Gradients of radial strain (barreling) were also observed at the lower confining pressures (not shown), e.g. at and below 34 MPa but surface cracking was not observed with confinement.

The results given in Figures 3, 4 and 5 indicate that the modulus, the flow stress, and the work hardening coefficient increase with increasing pressure. At 138 MPa and a strain rate of 0.001/sec. the estimated yield strengths of PBS 9501, LX-14, PBX 9501 and PAX 2A are less than but approach the value of aluminum (at 0.1

MPa) and are small but non trivial fractions of the value of steel (also at 0.1 MPa) (Handbook of chemistry and physics, 1945). In addition, the yield strengths of these composites have been found to increase significantly with increasing strain rate so that at the higher strain rates encountered in field use the yield strengths approach even closed to those of metals. Therefore, at the higher confining pressures used in this work these four composites have some metal-like properties, i.e., they fail by yield and plastic flow, exhibit work hardening, and the yield strengths of all four at the highest pressures and strain rates approach the values of metals. This behavior is to be contrasted with the sometimes brittle ceramic-like properties when these types of materials are unconfined (see Figures 2 and 6).

4. DISCUSSION

General considerations for a discussion of the pressure dependence of the stress-strain curves of composites include the following: a) the pressure dependence of the mechanical properties of the individual components of the composites; b) the effect of pressure on the interfaces of the composite; and c) the effect of pressure on defects such as voids and cracks (Hoppel et al., 1995). For the composites under consideration, the mechanical properties of interest here for the polymer component are expected to be a function of pressure while the same mechanical properties for the crystalline components, i.e., sucrose or HMX are expected to be insensitive or independent of pressure (Hoppel et al., 1995). A discussion of the pressure dependence of the modulus is followed by a discussion of the pressure dependence of the flow stress and the failure processes.

4.1 Modulus

There are several factors influencing the pressure dependence of the modulus including the following: a) finite elastic strains; b) collapse of voids; c) changes of the glass transition temperature; and d) the relative contributions from the binder and the explosive or sucrose. Because polymers are softer than many materials, the strains are larger and in many cases it is necessary to consider finite elastic strains rather than the more usual infinitesimal elastic strains. When this is done the modulus is found to increase linearly with pressure for the conditions of this work (Hoppel et al., 1995). The rate of increase with increasing pressure is dependent only on Poisson's ratio (at atmospheric pressure) and is between approximately three and eight. The initial slope of the curve of the modulus versus pressure for PBS9501 is about 400 (see Figure 3 and attendant discussion) so that finite elastic strains can only account for a small part of this initial slope. However, finite elastic strains may account for the slope at the highest pressures of Figure 3.

The collapse of voids and cracks may account for a part of the initial increase of the modulus with increasing pressure. The porosities of the undeformed PBS9501 samples are estimated to be between 2% and 2.4%, and the modulus is exponentially dependent on porosity for some types of porosity (Wang, 1984; Wiegand and Pinto, 1991). However, measurements of a group of PAX 2A samples pressed to a range of densities indicate a change of only about 22% in Young's modulus for a 4.2% change in porosity (Wiegand, D. A., unpublished results). In addition, data for a group of Composition B samples indicate a change of Young's modulus of 63% for a change of porosity of 2% (Wiegand and Pinto, 1991). These results suggest that only a part of the modulus increase for PBS9501 with pressure as given in Figure 3 can be associated with a decrease in porosity. It is also to be noted that for the collapse of porosity to account for a significant part of the initial slope, Figure 3, the pores must collapse at relatively low pressures compared to the much higher pressures used in preparation by pressing. Measurements at elevated pressures as a function of porosity would be useful in determining the role of porosity in the pressure dependence of the modulus.

An increase in the glass transition temperatures (T_g) with pressure can also cause very significant increases of the modulus under appropriate circumstances (Hoppel et al., 1995; Patterson, 1964). Since the T_g 's of most of the composites considered here are below the measurement temperature (see Table 1), increases in T_g will result in increases in the modulus (Wiegand, 1995). Measurements of the modulus as a function of temperature in the vicinity of the T_g would be helpful in determining the magnitudes of increases that could be expected. Of course determining the pressure dependence of T_g would be especially valuable in resolving this matter.

As noted above it is also necessary to consider the relative contributions to the modulus by the binder and by the explosive or sucrose as a function of pressure. The total strain can be considered to result from displacements in the binder and displacements in the explosive or sucrose. The component of the total displacement due to the explosive or sucrose is expected to be independent of or insensitive to pressure. However, the component of the total displacement due to the polymer in the binder is expected to decrease with increasing pressure since the modulus of polymers has been found to increase with increasing pressure (Hoppel et al., 1995). Thus, the total displacement and so the total strain is expected to decrease with increasing pressure. Hence the modulus of the composite is expected to increase with increasing pressure as observed. The temperature and strain rate dependence of Young's modulus at 0.1 MPa indicates that the polymer in the binder plays a very significant role in determining Young's modulus at this pressure (Wiegand,

1995). However, as pressure is increased and the component of the total displacement due to the binder decreases, the component of the total displacement due to the explosive will become more significant. Thus, at higher pressures the modulus is also expected to be less sensitive to pressure as observed. Therefore, the observed increase of the modulus with increasing pressure and, in addition, the observed decrease in sensitivity of the modulus to pressure with increasing pressure may both be due at least in part to a decrease in the component of the total displacement due to the binder as pressure is increased. Measurements as a function of temperature and strain rate at elevated pressures may be useful in determining the importance of these processes in determining the pressure dependence of the modulus.

In summary, the increase of the modulus with increasing pressure may be due to several factors, i.e., finite elastic strains, the collapse of porosity, an increase of T_g , and a decreasing contribution of the polymer binder to the total displacement. Additional work is clearly indicated to determine the roles of the mechanisms considered in the increase of the modulus with increasing pressure.

4.2 Flow Stress

At atmospheric pressure (0.1 MPa) it is clear that crack processes take place during compression (see Figure 7). It is also probable that some plastic flow occurs because of the shape and condition of the samples after deformation. It is to be noted that crack growth and plastic flow need not take place in the same part of a composite. For example, cracks may be primarily interfacial while plastic flow may take place primarily within one of the components of the composite, e.g., the binder. With increasing pressure the results suggest that crack growth is decreased and that plastic flow is increased. In particular, surface cracking which is observed without confinement is not observed when samples are confined (see Figure 7). In addition, it is found that the fractional density changes on deformation under confinement are very small compared to the fractional changes in dimensions, thus suggesting that deformation takes place at approximately constant volume.

It is expected that the stress required for crack growth will increase with pressure because of the observed increase of the modulus. There may also be an increase in the effective surface energy because of the additional work that must be done against the confining forces to create new internal crack surface area. This will also cause an increase in the stress required for crack growth. Thus, pressure inhibits crack growth and so the observed lack of surface cracking at elevated confining pressures and the shift to deformation by plastic flow is not

unexpected. Therefore, there is a brittle-like to ductile transition as pressure is increased. Many brittle materials become ductile under hydrostatic pressure and a similar transition has been observed in other materials (Wiegand et al., 1991; Pinto and Wiegand, 1991; Hoppel et al., 1995).

The initial increase of the flow stress with increasing pressure at low pressures is, therefore, most probably due in part to an increase of the stress required for crack growth with increasing pressure. However, this increase may also be due in part to the transition of the failure mechanism from primarily crack processes to primarily plastic flow. The increase of the flow stress with increasing pressure may also be due to some of the same reasons as the increase in the modulus. The yield strength has been found to increase exponentially as porosity decreases in the same manner as the Young's modulus (Wiegand and Pinto, 1991; Knudsen, 1959). The flow stress considered here is expected to have the same porosity dependence. In addition, the flow stress may increase with an increase in the glass transition temperature since the failure strength increases as temperature is decreased in the vicinity of the glass transition temperature at atmospheric pressure (Wiegand, 1998). And finally yield in the explosive or sucrose may become more significant as the yield strength of the binder increases as pressure is increased, thus accounting in part for the decreased sensitivity of the flow stress to pressure at higher pressures. Some of the yield relationships developed for polymers as a function of pressure may describe the pressure dependence of the flow stress observed in this work (Hoppel et al., 1995).

In summary, the same mechanisms which may determine the pressure dependence of the modulus may in part determine the pressure dependence of the flow stress. However, the pressure dependence of cracking appears to play a significant role in the pressure dependence of the flow stress at least at the lower pressures. Microscopic studies of deformed samples as a function of pressure during deformation should be helpful in determining the importance of crack processes.

5. SUMMARY

The results indicate significant increases of the modulus, the flow stress, and the work hardening coefficient with increasing pressure and the sensitivity of all three of these quantities to pressure decreases markedly with increasing pressure. The pressure dependence of the modulus is discussed in terms of several factors including the following: changes in porosity, a shift of the glass transition temperature, changes in the relative contributions of the binder and the explosive/sucrose to the modulus, and the effect of finite strains, all as a function of pressure. The pressure

dependence of the flow stress may be due to some of these same factors, but is also influenced by the effect of pressure on crack growth processes.

ACKNOWLEDGEMENTS

The authors are indebted to D. Idar and B. Asay for providing the samples of PBS 9501 used in this work

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